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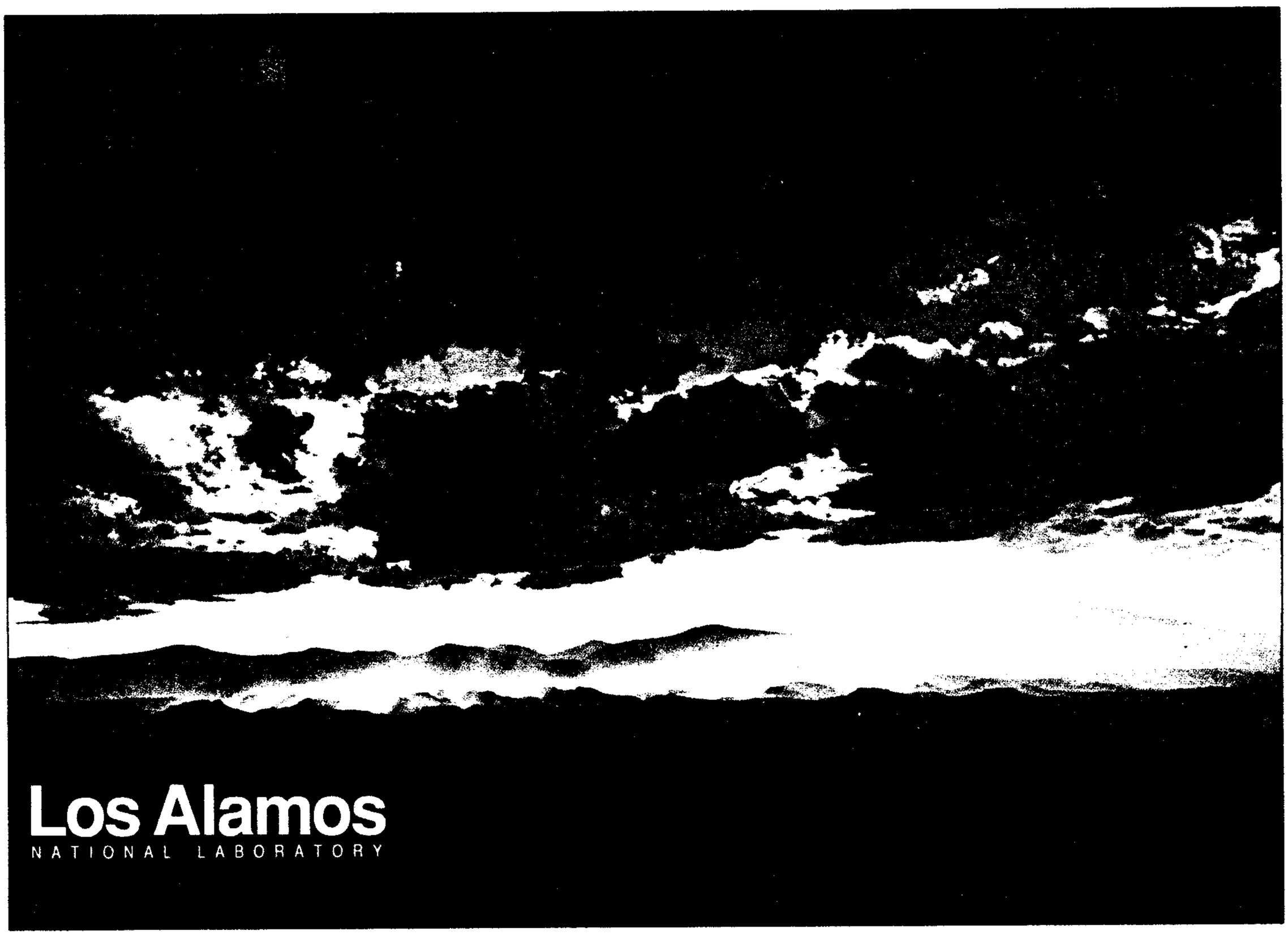

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ABSTRACT

In the fall of 1994, Rocky Flats Environmental Technology Site (RFETS) committed to offering approximately one metric ton of excess weapons plutonium for safeguards verification by the International Atomic Energy Agency (IAEA). At that time RFETS decided to offer cans of plutonium oxide of varying purity, stored one or two to a package in 10-gal. drums, and contracted with Los Alamos to obtain a multiplicity counter to measure these materials. A multiplicity counter was deemed necessary because of IAEA experience that the standard coincidence counting techniques routinely used in IAEA inspections would fail for plutonium-bearing materials of variable purity. RFETS also thought that such an instrument would be useful for their domestic safeguards needs.

The 30-Gallon-Drum Neutron Multiplicity Counter (NMC) was shipped to RFETS and installed early in 1995. The counter was characterized with ^{252}Cf sources before leaving Los Alamos, and this characterization was later corrected to plutonium through measurements made with an identical instrument. Several potential measurement problems were investigated with the 30-Gallon-Drum NMC before the IAEA's initial physical inventory verificationat RFETS in December 1995. These investigations were motivated by a concern that the neutron multiplicity counting technique had never been applied to samples packaged in the manner of those offered for IAEA inspection. Selected drums were measured in the 30-Gallon-Drum NMC, and the results were compared to site inventory values derived from calorimeter measurements and gamma-ray isotopic measurements analyzed by GRPAUT or its TRIFID counterpart, EPICS. Los Alamos helped RFETS interpret the multiplicity counting results relative to these data.

In this paper we will present assay data for NMC measurements of over 100 10-gal. drums, most of which were verified by the IAEA in December 1995 at RFETS, and discuss the 30-Gallon-Drum NMC's accuracy and precision relative to site inventory values. We will discuss the expected precision in both the neutron multiplicity assays and the site inventory values and how neutron counting precision and isotopic measurement precision contribute to the overall precision of the assays obtained with this instrument.

INTRODUCTION

The 30-Gallon-Drum Neutron Multiplicity Counter (NMC) [1] shown in Fig. 1 was originally designed to investigate the use of multiplicity counting to assay bulk plutonium stored in 30-gal. drums. The primary goal[2] of its design was to provide a measurement precision based on neutron counting statistics of between 1 and 3% in 30 minutes for large mass materials whose (alpha, n) neutron emissions are from zero to one times their spontaneous fission neutron emissions. The application of this instrument to materials at Rocky Flats Environmental Technology Site (RFETS) was necessitated by the 10-gal.-drum packaging and the fact that no other multiplicity instrument existed that could accommodate a package this large. Although the 30-Gallon-Drum NMC was not specifically designed for this application, we felt confident it would adequately meet the Initial Physical Inventory Verification (IPIV) requirements of the International Atomic Energy Agency (IAEA).

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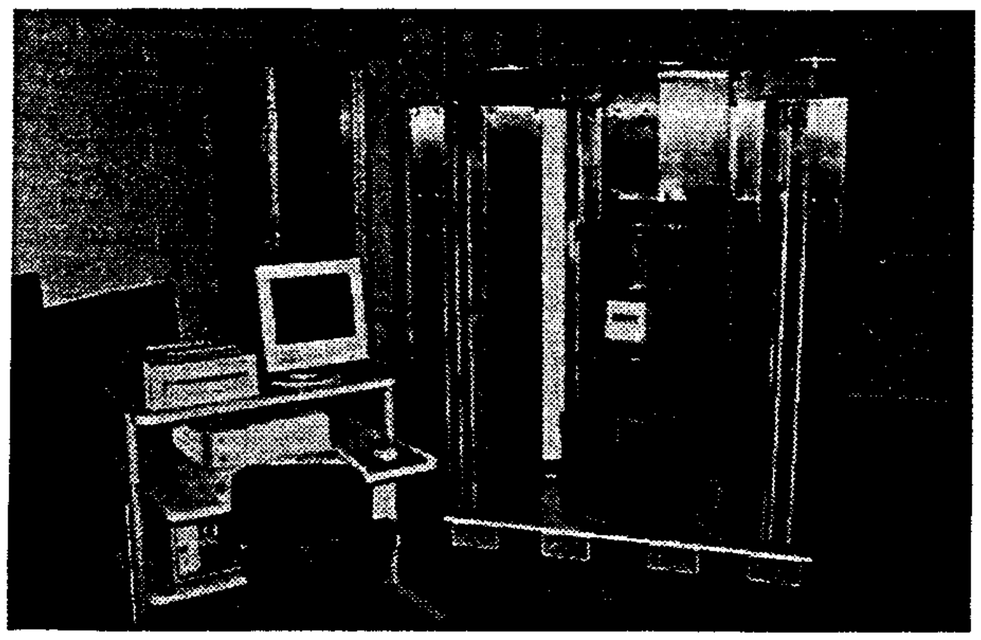

Fig. 1. The RFETS 30-Gallon-Drum Neutron Multiplicity Counter.

Before the IAEA's IPIV in December of 1995, a small number of drums were measured to verify the system's performance for materials similar to those to be offered for IAEA safeguards. During these initial measurements we deduced that the instrument was performing up to specifications and that the unusual stacked arrangement of the inner oxide cans in the 10-gal. drums, shown schematically in Fig. 2, would not cause difficulty for the multiplicity algorithms. These initial data, together with the IPIV measurements, form a large database of measurements of drums that are both within and not within the excess materials offering. This database is of adequate size to well assess the 30-Gallon Drum NMC's performance relative to both measurement precision and accuracy.

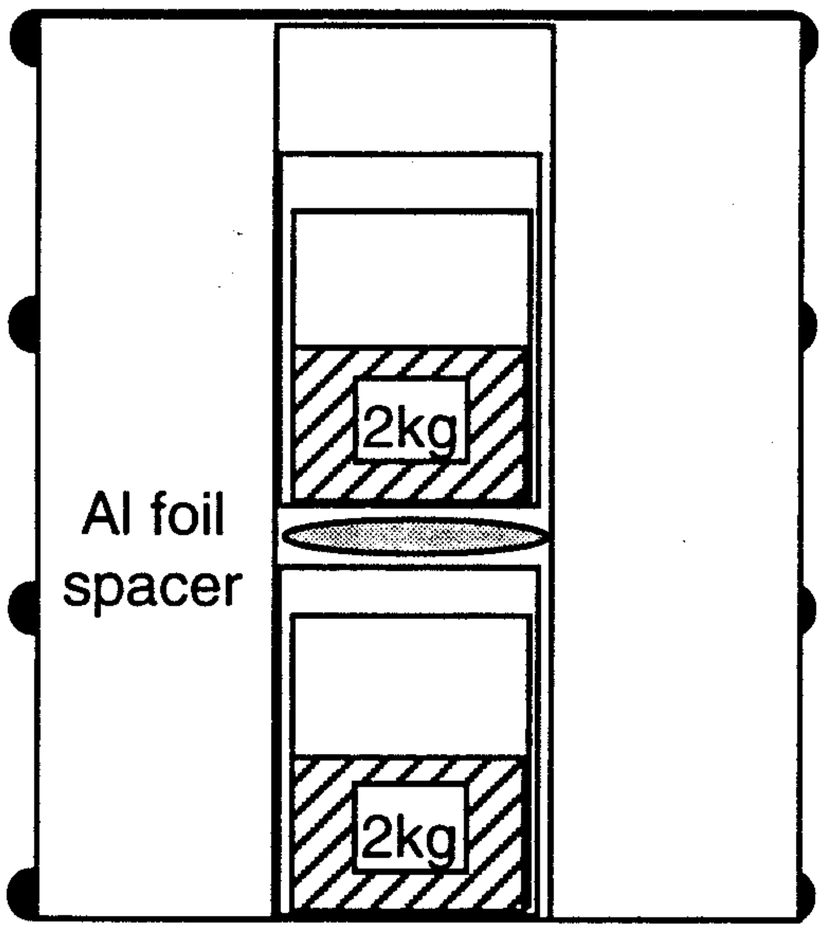

Fig. 2. Nominal RFETS drum loading; variations in sample masses exist. Drum is approximately 44 cm high by 38 cm wide.

THE MEASUREMENT DATABASE

Both during the IPIV and before, a variety of count times were used for the measurements made with the 30-Gallon-Drum NMC. The majority of the measurements had 30-minute count times, however. For this reason, the data chosen to assess the performance of the 30-Gallon-Drum NMC were limited to those samples counted for 30 minutes or more. The data from count times longer than 30 minutes were truncated to the first 30 minutes of collection so that conclusions about measurement precision could be based on a uniform counting time across all retained measurements. Using these criteria, the database consists of 106 measurements. To assess measurement accuracy, the assay results were compared to site inventory declarations for the individual cans in each drum. These declared values were originally obtained by calorimetry and gamma-ray isotopic measurements analyzed by GRPAUT[3] or its TRIFID[4] counterpart, EPICS, between 1985 and 1990. From the combination of gamma-ray isotopic and calorimeter measurements, the total plutonium mass of each can in a drum is believed to be known to about 0.5%. The ratio of the ^{240}Pu-effective mass (what a neutron multiplicity counter measures) to the total plutonium mass is only known from 2 to 6%. The plutonium oxide drums in this sample population contain an average total plutonium mass of 3.4 kg but range from 0.7 to 4.1 kg. The average ^{240}Pu-effective mass is 6.0% of the total plutonium mass, and this ratio varies only slightly from 5.8% to 6.6%.

The isotopic ratios for each can in a drum were not necessarily identical. The multiplicity software takes this into account by computing a composite isotopic from the declarations for each can. Gamma-ray measurements made during the IPIV revealed that the declared position of an individual can in a drum was not always accurate. However, an advantage of this multiplicity counter and the neutron counting technique itself is that they are insensitive to the position of an individual can in the drum as long as the drums are placed consistently with respect to position in the sample chamber.

ASSAY RESULTS

Table I gives the average assay results obtained with the 30-Gallon-Drum NMC compared to site declared values for these 106 samples. The data were analyzed three ways to obtain an effective ^{240}Pu mass: a neutron multiplicity analysis and two standard neutron coincidence techniques, the "Known alpha" and the "Known M" methods. In the "Known-alpha" method,[5] alpha, the ratio of (alpha,n)

TABLE I. Assay Method Comparison

Assay Method	Known-Alpha	Known-M	Multiplicity (All Samples)	Multiplicity (Alpha Less Than 3.1)
Number of Samples (N)	106	106	106	98
Average [Declared (D) - Assay (A)] / Assay (%)	-40.9	-0.80	0.62	0.58
1 sigma	65.2	8.09	4.18	3.54
1 sigma / $\sqrt{N}$	6.3	0.79	0.41	0.36
Minimum [(D-A)/A] (%)	-366.0	-27.7	-14.0	-8.1
Maximum [(D-A)/A] (%)	4.4	14.8	13.5	13.5
% Within ± 3%	13.2	29.2	61.3	65.2
% Within ± 18%	48.1	94.3	100.0	100.0

neutrons to spontaneous fission neutrons emitted by a sample, is calculated using the declared isotopic ratios. This calculated alpha is then combined with the measured total neutron rate and the coincidence rate to deduce an assay. In the "Known-M" method,[6] sample self-multiplication is related to the fissile mass, and the total and coincidence neutron rates are combined with this information to deduce an assay. For this analysis the self-multiplication, M, from the multiplicity measurements was used to define the relationship between the ^{239}Pu-effective mass and M. In the multiplicity analysis a third measured quantity, the number of coincident triples, is added to the analysis and no assumptions about the sample are made. For all three methods, sample isotopic information is necessary to compute the total plutonium mass from the ^{240}Pu-effective mass measurement so a comparison could be made to site inventory values.

The Known-alpha method (Fig. 3) is a standard technique used by the IAEA to measure plutonium oxide but clearly does very poorly for these impure samples. The Known-M technique (Fig. 4) does surprisingly well, probably due to the constrained geometry of the individual samples in the drums and the more or less constant mass loading. The multiplicity technique, however, improves the average agreement between the declared values and the assay by nearly a factor of two from the best two-parameter assay. Both the multiplicity technique and the Known-M method produce statistically unbiased results relative to the site inventory values and show no trends as a function of mass, self-multiplication, or the ratio, alpha.

Included in Table I are the percentage of this sample population that agreed to within plus and minus 3% and 18% of the declared values. Only the multiplicity technique provides an adequate rate of success for a 3% limit and 100% success for the 18% limit. These rejection

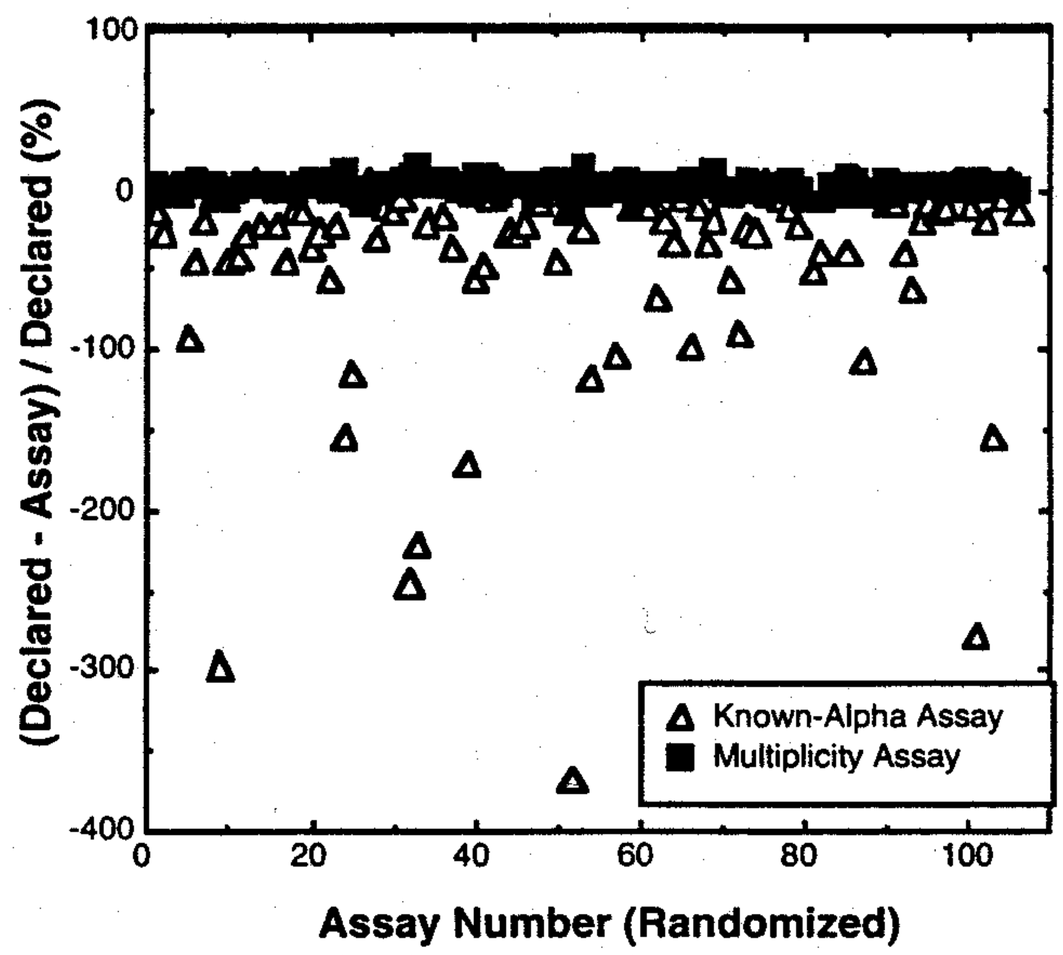

Fig. 3. Comparison between "Known-Alpha" assay results and multiplicity results.

limits, which correspond to 3 standard deviations, are based on expected measurement accuracies of 1% and 6%, respectively, and are consistent with IAEA practices related to bias and partial-defect tests for plutonium inventory verifications.

Figure 5 shows the multiplicity assay results relative to site declared mass vs a randomized assay number. Included in this figure is an estimate of assay precision. The assay precision was estimated based on counting statistics and an estimate of the random error in the isotopic ratios that are needed to convert the measured fertile mass to total plutonium mass. The assay precision based on counting statistics was estimated by counting each sample for 60 separate 30-second intervals and computing the standard deviation in the mean assay result. For this population of 106 samples the average relative assay precision due to

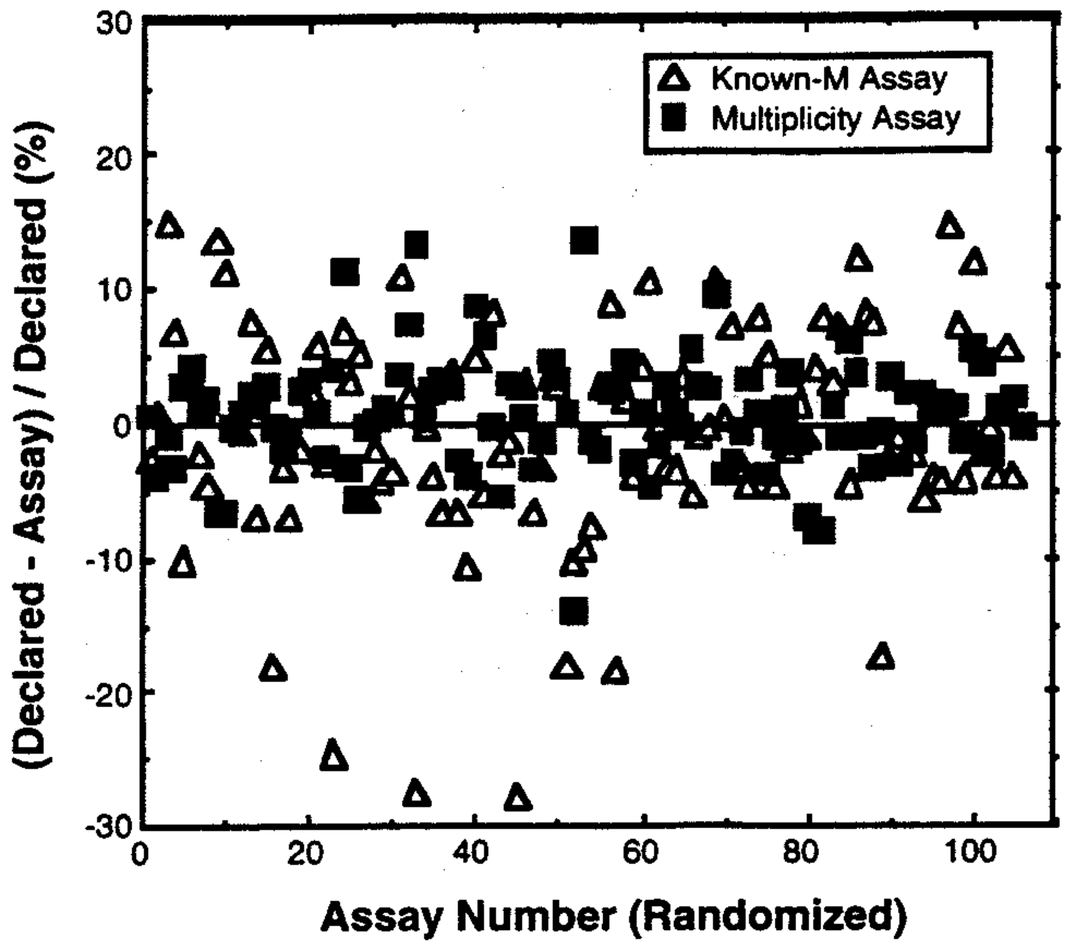

Fig. 4. Comparison between "Known-M" assay results and multiplicity results.

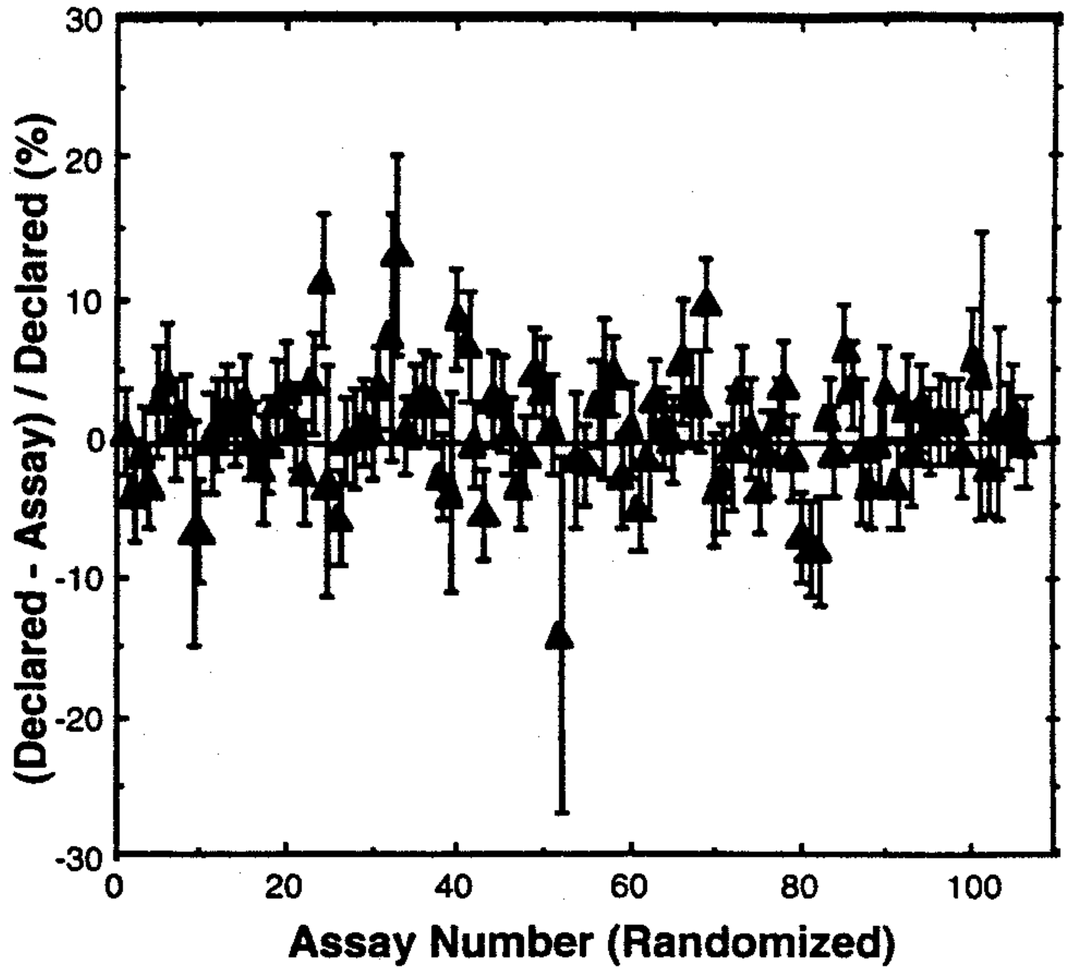

Fig. 5. Multiplicity assay results with an estimate of precision.

neutron counting statistics alone was 2.6%. The best precision was 0.9%; the worst was 12.6%. To account for the random error in the isotopic measurement, because the estimated isotopic precision was not available for all samples, the average precision due to isotopic measurements was obtained for a representative 10% of the population. For this sampling, the average relative precision in the ratio of fertile mass to total plutonium mass was 2.6% but ranged from 2.4% to 6.0%.

For all 106 samples, the average precision estimate for an assay compared to the declared value was 3.8% and ranged from 2.8% to 12.9%. For errors calculated in this manner, only one assay out of the population of 106 failed to agree to within 3 standard deviations of the declared value. This particular sample had poorer isotopic precision than the average, however. When its correct isotopic precision was used, it also fell within 3 standard deviations of the declared value.

In neutron coincidence counting, assay precision is dependent on the total neutron rate that in turn depends on the sample mass; self-multiplication; and ratio, alpha. For this sample population, the plutonium mass of the samples is fairly uniform, and the self-multiplication of neutrons varies relatively little from drum to drum because of the uniform geometry and nearly uniform mass loading. In fact, the average multiplication yielded from the multiplicity algorithms for this population was 1.177 with a sigma of only 0.033. However, the ratio, alpha, varies much more dramatically. Figure 6 shows the effect of this ratio on the estimated multiplicity assay precision due to counting statistics. For pure plutonium oxide

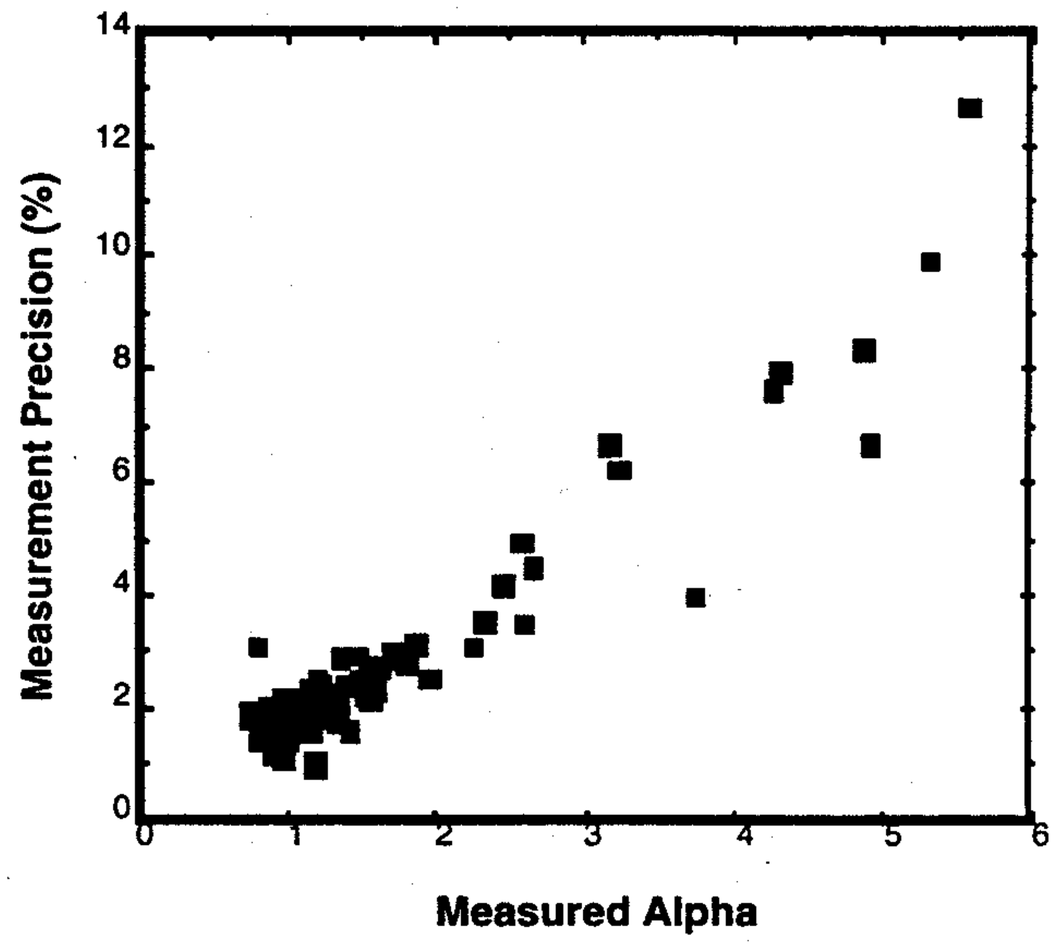

Fig. 6. Estimated assay precision due to counting statistics vs "alpha" from the multiplicity assay results.

having these isotopic ratios, alpha would vary only from 0.76 to 0.91. More than 80% of these samples have measured alpha ratios greater than this. Figure 6 shows that as alpha increases the counting precision becomes poor. To compensate for this effect, samples with large alphas need to be counted longer to achieve better precision. For samples having an average ^{240}Pu-effective isotopic ratio precision of 2.6%, an average assay precision due to counting statistics of 5.4% is needed for the 1σ in the assay to be 6%. Eight samples in this population had alphas such that their estimated counting precision was higher than 5.4%. All had alphas greater

than 3.1. Thus for samples with these high alphas the count time should be increased to achieve an overall assay precision of 6%. In Table I we include a column for samples limited to alphas less than 3.1. In Table II we summarize the neutron counting statistics for the population.

In Fig. 7, we show the predicted precision of this instrument calculated by Ensslin's figure-of-merit code[7] for alphas from 1 to 5 and a 30-minute count time. In this population of samples, the average ^{240}Pu-effective mass was about 207 grams with a sigma of 42 grams. The average alpha ratio from the multiplicity algorithms for this population was 1.5 with a sigma of 1.0. For the average mass, self-multiplication, and alpha, Ensslin's code predicts an assay precision due to neutron counting precision of 2.9% that agrees very well with the average counting precision obtained from the measurements of 2.6%. Overall, the counting precision obtained from this population agrees well with the predictions of Ensslin's code.

THE EFFECT OF COUNT TIME

A major advantage of neutron counting is that an assay can be performed rapidly, and a large throughput is possible. From the above, an overall assay precision of 6% or less was achieved for 92.5% of this population with a 30-minute count time. For this segment of the population with alpha less than 3.1, one standard deviation for the assays compared to the declared value was 3.5%. To test the effect of count time on assay precision, the neutron counting data for this population was segmented into 15 and 20-minute count times and assays deduced. For the 20-minute count time, standard deviation for the assays compared to the declared value increases to 4.0%, and for the 15-minute count time it increases to 4.7%. All samples for both count times still fall within plus or minus 18% of the declared values and 62% fall within plus or minus 3% for the 20-minute count time. Only one additional sample failed to fall within plus or minus 3% when the count time was decreased to 15-minutes. From this we can conclude that shorter count times are acceptable for many of these samples, particularly those with smaller alphas. However, for the whole population, the 30-minute count time gives the best probability for success.

ROW RATIOS

The 30-Gallon-Drum NMC has three rows of tubes to attain a neutron detection efficiency high enough for multiplicity counting to be successful. The innermost row is the most sensitive to low-energy neutrons; the outermost row is most sensitive to high-energy neutrons. The ratio of the total neutron rates in these rows provides a measure of the mean energy of detected neutrons. Figure 8 shows this ratio for this set of measurements. The shaded area in the figure is the range of ring ratios obtained at Los Alamos during measurements of pure plutonium oxide materials made with the identical twin of the RFETS NMC. The vast majority of these samples contain impurities that change the energy signature relative to pure plutonium oxide. Samples with ratios smaller than the range for pure material have mean neutron energies higher than that for pure plutonium oxide. Samples with higher ratios have mean energies that are lower. From this we can conclude that these samples contain a variety of different low Z impurities. However, the 30-Gallon-Drum NMC's performance does not appear to be affected in a statistically significant way by these energy shifts.

CONCLUSIONS

The 30-Gallon-Drum NMC at RFETS has been used successfully at RFETS to measure excess plutonium oxide. The instrument has performed within specifications for samples containing a large variety of impurities. All of the samples assayed using the multiplicity counting technique agreed to within plus or minus 14% of the site inventory values; 61% fell within plus or minus 3%. The average neutron counting precision was 2.6% for the 30-minute count time but varied dramatically with alpha. Longer count times are desirable for samples having

TABLE II. Assay Precision Based on Neutron Counting Statistics Alone for 30-Minute Count Times

	All Samples	Samples with Alpha Less Than 3.1
Average Precision (%)	2.6	2.1
Standard Deviation (%)	1.8	0.7
Best Precision (%)	0.9	0.9
Worst Precision (%)	12.6	4.9

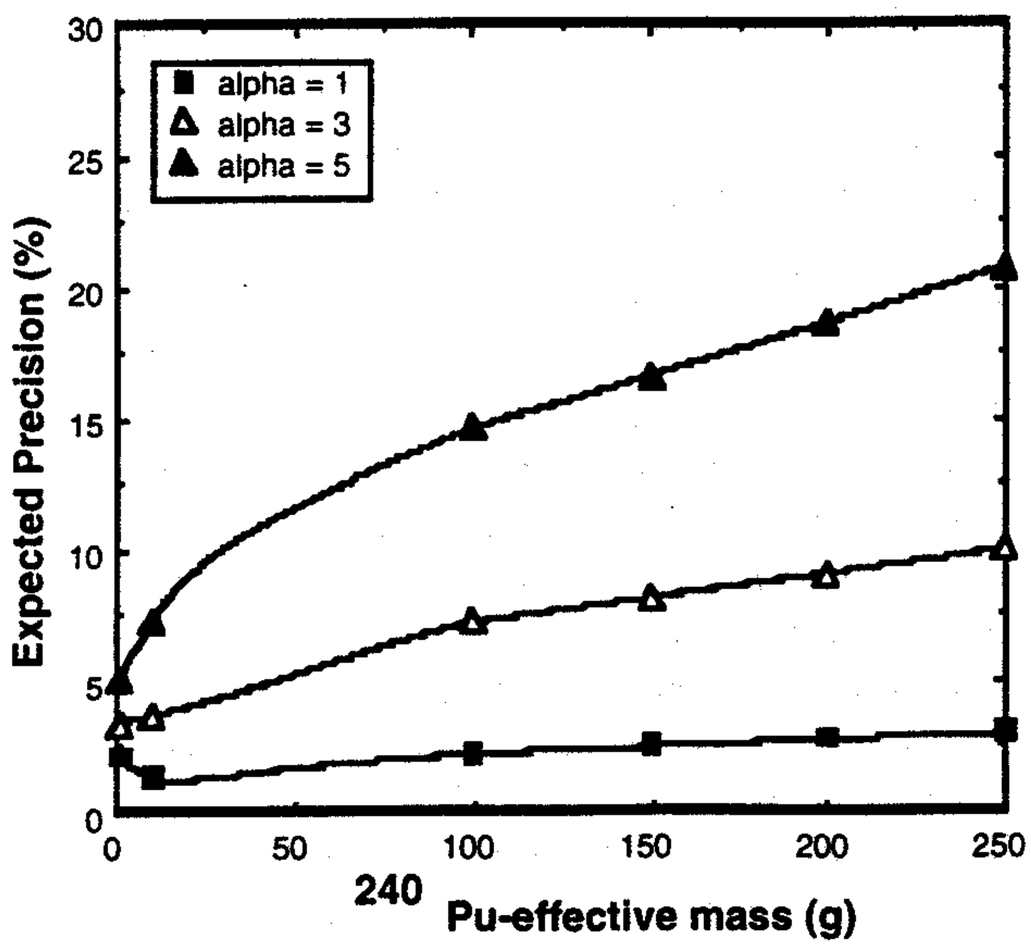

Fig. 7. Expected assay precision due to counting statistics for the RFETS 30-gal. Drum NMC from Ensslin's figure-of-merit code.

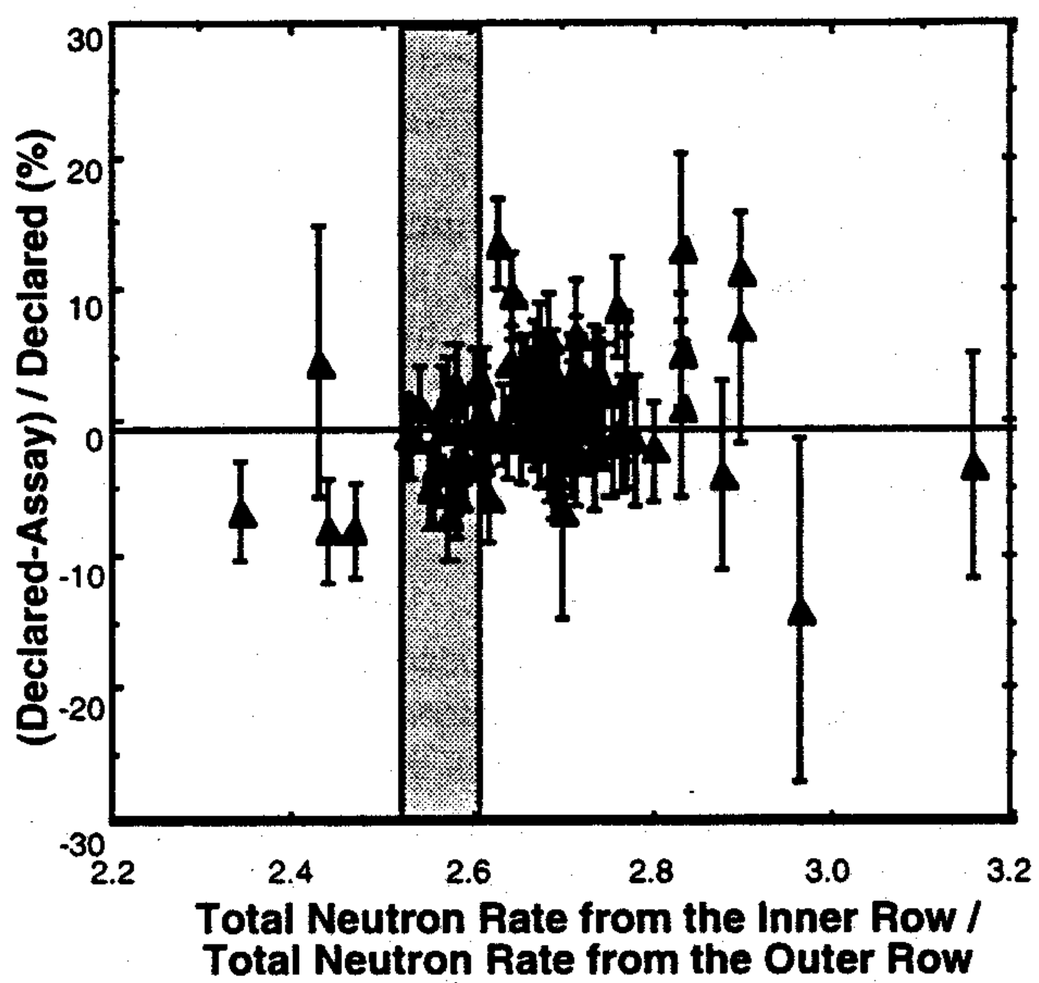

Fig. 8. Neutron rate "row ratios" showing the effect of impurities on the mean neutron energy. The shaded area is the range for pure plutonium oxide.

alphas above 3.1 to improve neutron counting precision. Only 7.5% of the samples in this population had alphas above 3.1. The 1σ agreement between the multiplicity counter and site declarations was 4.2% for all samples measured and 3.7% for samples having alphas below 3.1.

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